REMARKS

Favorable reconsideration is respectfully requested.

The claims 1-3, 6 and 7.

The above amendment is responsive to points set for in the Official Action.

With regard to Official Action paragraph 2, in the specification, the pressure is given in bar; see Table 1. However, at page 7 it is mentioned that the oxygen partial pressure <u>difference</u> can be 6-7 decades or more across the membrane. In this connection decade means: order of magnitude, which definition has been inserted in the specification. No new matter is involved. In this regard, see the definition of "decade" in Webster's Third New International Dictionary, page 583 (1971), attached.

Claims 1-3, 6 and 7 have been rejected under 35 U.S.C. 102(b) as anticipated by the disclosure of either one of the following references as applied in the Norwegian Search Report: (1) Mazanec et al. (5,306,411) or (2) Thorogood et al. (5,240,480) or (3) Mazanec et al. (5,714,091) or (4) EP 0,438,902 or (5) NO 306014.

This rejection is respectfully traversed.

U.S. 5,306,411 (1) describes a membrane formed with a perovskite. Said membrane, however, does not include Al (aluminum) in contrast to the membrane according to the present invention. The membrane according to the present invention is represented by the formula as shown in claim 1 where y' > 0, i.e. the membrane shall include aluminum.

The membrane according to the present invention is also not disclosed or suggested by US 5,240,480 (2). According to (2) the ratio between the A-type and B-type cations shall be 1, while said ratio shall be greater than 1 for the membrane according to the present invention; i.e. v/w > 1, while in (2) x + x' + x" = 1 and y + y' + y" = 1. Please note page 11, line 31 - page 12, line 21 in the present specification where it is explained why it is important that said ratio be greater than 1.

Similarly, this is also true for the material described in US 5,714,091 (3). Said ratio shall be 1. Furthermore, the material disclosed in (3) does not include aluminum.

The material disclosed in EP 438902 (4) and NO 306014 (5) is identical to the material disclosed in (1) and (3), respectively. Hence, no further comments are necessary for these two references.

Claims 1-3, 6 and 7 have been rejected under 35 U.S.C. 102(b) as being clearly anticipated by the disclosure of (6) Cable et al. (US 5,910,238), specifically at Table 1.

This rejection is respectfully traversed.

US 5,910,238 (6) describes mixed metal oxides represented by the formula

$$(A_{1-a}A'_a)(B_bB'_{1-b})O_{3-c}$$
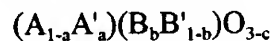
consisting of a wide range of elements and compositions. Strictly speaking, the nomenclature of this formula restricts compositions to those having a 1:1 ratio of A-type and B-type cations, since $1 - a + a = 1 - b + b = 1$. A, A', B and B' clearly represent elements, but are apparently also confused with numbers. Presumably, $0.9 \leq (A + A')/(B + B') \leq 1.1$, which therefore strictly speaking is meaningless, indicates a range in the A-type to B-type cation ratio.

The compositions claimed in (6) encompass a total number of 36 elements. The formula describes hundreds, if not thousands, of mixed metal oxides described in the literature prior to the date of filing.

Given the general nature of the formula of reference (6), it is true that the formula encompasses the compositions described by the present claim 1. However, to translate this formula to the compositions described by the present claim 1, the following restrictions have to be made:

- A contains not a mixture of cations, but only one.

- A' contains not a mixture of cations, but only one.

- $b = 0$ (i.e. B is absent).

- B' contains not less than two cations and not more than three.

- $0 < a < 1$.

After applying these restrictions, A may still represent any one of 19 elements, A' any one of 6 elements, and B' any combination of two or three from a selection of 10 elements. Hence, applying all these restrictions to the general formula of (6), a total of $19 \times 6 \times (45 + 120) = 18810$ combinations remain. Of these 18810 combinations only two coincide with the compositions of present claim 1.

It is common knowledge that among the countless compositions encompassed by the general formula of (6), there are countless compositions that may be used to transport oxygen in a laboratory experiment. However, confronted with the task of identifying a composition for use in an <u>industrial</u> application, the challenge is of a different nature than merely picking any of these countless compositions. The present patent application claims specific compositions as a result of demonstrating in a detailed and thorough manner the advantages of certain compositional features. This demonstration shows why the vast majority of compounds encompassed by a general formula like the one given in (6) are not useful as oxygen membranes.

One of the most important and novel features of the present invention is the demonstration of the importance of the A/B cation ratio of the perovskite $A_A B_B O_3$. Normally this ratio equals one. (6) claims compositions with $0.9 \leq (A + A')/(B + B') \leq 1.1$. However, according to Table 1 of (6), preferably $0.99 \leq (A + A')/(B + B') \leq 1.01$. Hence, an intention to deviate from the normal ratio of 1 is not demonstrated by this reference. The present invention, however, demonstrates that mixed metal oxide perovskites with A/B cation ratios equal to one will be unstable to kinetic decomposition, while those with ratios higher than one will be stable. The A/B ratio therefore will have a strong influence on the lifetime of a membrane.

Thus, reference (6), with its countless compositions, does not anticipate or suggest the specific composition of the present application. In (6) no information or knowledge regarding properties of compositions is communicated, other than a categorization of compositions into, on one hand "electronic conductors", and on the other "ionic and mixed ionic/electronic conductors". In fact, the compositions of the general formula encompassing the compositions of present claim 1, are listed as <u>electronic conductors</u> in Table 1 of (6). In sum, (6) does not anticipate or suggest the composition of present claim 1 as an oxygen membrane. Therefore, given the information in (6), the composition of present claim 1 would not be expected to be useful as an oxygen membrane but at best, an electronic conductor.

For the foregoing reasons, it is apparent that the rejection on prior art is untenable and should be withdrawn.

No further issues remaining, allowance of this application is respectfully requested.

If the Examiner has any comments or proposals for expediting prosecution, please contact undersigned at the telephone number below.

Respectfully submitted,

Turid RISDAL et al.

By: _Matthew Jacob_

Matthew Jacob
Registration No. 25,154
Attorney for Applicants

MJ/abm
Washington, D.C. 20006-1021
Telephone (202) 721-8200
Facsimile (202) 721-8250
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are inherently unstable as perovskites and that a great number of compositions encompassed by WO97/41060 are inherently unstable as brownmillerites under all conditions relevant to membrane processes. Furthermore, a large number of the compositions encompassed by US 5,306,411, US 5,712,220, and WO97/41060 are characterised by low or zero oxygen flux under all conditions relevant to membrane processes.

The main object of the present invention was to arrive at an improved membrane showing good stability against reaction with carbon dioxide and against reduction of oxide components to metal.

Another object of the present invention was to arrive at an improved membrane showing stability against kinetic decomposition and resistance to mechanical failure due to chemical expansion stresses.

The inventors found that a certain class of multicomponent metallic oxides are particularly suited as membrane materials in processes in which the membrane is subjected to a large potential gradient, e.g. oxygen partial pressure difference of 6-7 ~~decades~~ orders of magnitude or more across the membrane. These compositions overcome problems associated with kinetic decomposition. Additionally, due to their low chemical expansion and high stability against carbon dioxide and water, these materials are particularly suited as membranes for the production of syngas and for fossil energy conversion with economical CO_2 abatement.

The compositions according to the present invention are based on the so called perovskite structure, named after the mineral perovskite, $CaTiO_3$, but the cation stoichiometry is different from the ideal perovskite, and it is this difference that gives the compositions according to the present invention superior stability in a potential gradient. Furthermore, the process conditions associated with the production of syngas or fossil energy conversion with economical CO_2 abatement limit the selection of elements of which the perovskite membrane can consist.

Following the discussion and further limitations hitherto, the membrane material possessing the perovskite structure for use in said processes, must have a composition represented by the formula:

$$(La_{1-x}Ca_x)_v(Fe_{1-y-y'}Ti_yAl_{y'})_wO_{3-d}$$
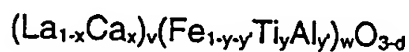
wherein x, y, y', v, w, and d each represent a number such that $0 \leq x \leq 1$, $0 \leq y < 1$, $0 \leq y' < 1$, $0 < (y+y') < 1$, $0.9 \leq v \leq 1$, $0.9 \leq w \leq 1$, and d equals a number that renders the compound charge neutral and is not less than zero and not greater than about 0.8, and preferably $0.95 \leq v \leq 1$ and $0.95 \leq w \leq 1$.

Stoichiometric perovskite compositions represented by said formula, i.e. $v = w = 1$, are kinetically unstable when subjected to large gradients (6-7 ~~decades~~ *orders of magnitude* or more) in the oxygen partial pressure. The kinetic decomposition that occurs in these materials gives rise to the formation of decomposition products on at least one of the membrane surfaces and a decrease in the oxygen flux with time. Such kinetic decomposition in the stoichiometric perovskite materials is exemplified by the present examples 12 and 15 and Figures 4, 8, 9, and 10. Kinetic decomposition becomes more pronounced when w > v. Therefore, stoichiometric perovskites (v = w), or perovskites with A-site deficiency (w > v) represented by said formula can not be used as membranes in said processes.

The exclusion of stoichiometric and A-site deficient perovskites, excludes the use of the compositions of US 5,712,220 and WO97/41060, and excludes the use of the preferred compositions of US 5,306,411 referenced in the "Background of the invention", as membranes in the said processes. Said compositions of US 5,712,220, US 5,306,411, and WO97/41060, are expected to decompose in the large oxygen partial pressure gradient of said two processes, resulting in decreasingly poor oxygen permeation and eventually to cracking and complete breakdown of the membrane.

EXAMPLE 24

### OXYGEN PERMEATION TEST OF A $La_{0.05}Ca_{0.95}Fe_{0.145}Ti_{0.825}O_{3-d}$ MEMBRANE

An oxygen permeation test is conducted according to the procedure described in Example 11, except a membrane disk of a composition represented by the formula $La_{0.05}Ca_{0.95}Fe_{0.145}Ti_{0.825}O_{3-d}$ is used. Fig. 14 shows a ternary diagram of the system $LaFe_wO_{3-d}$-$CaTi_wO_{3-d}$-$CaFe_wO_{3-d}$. The composition of the material is located near the $CaTi_wO_{3-d}$ apex of the ternary system, at the point marked "B". Low electronic conductivity, especially at low oxygen partial pressures is characteristic of materials with compositions in this region, and a low oxygen flux is measured.

EXAMPLE 25

OXYGEN PERMEATION TEST OF A $La_{0.25}Ca_{0.75}Fe_{0.63}Ti_{0.34}O_{3-d}$ MEMBRANE

An oxygen permeation test is conducted according to the procedure described in Example 11, except a membrane disk of a composition represented by the formula $La_{0.25}Ca_{0.75}Fe_{0.63}Ti_{0.34}O_{3-d}$ is used. Fig. 14 shows a ternary diagram of the system $LaFe_wO_{3-d}$-$CaTi_wO_{3-d}$-$CaFe_wO_{3-d}$. The composition of the material is located near the centre of the ternary system, at the point marked "C". Ordering of oxygen vacancies and cations is characteristic of materials within the system with compositions with $CaFe_wO_{3-d}$ content higher than a limit depending on the temperature and oxygen partial pressure. The obtained oxygen flux decreases with time as the ordered phase forms.

These Examples demonstrate that the oxygen separation membranes of the present invention are particularly suitable as membrane materials in processes in which the membrane is subjected to a large potential gradient, e.g. oxygen partial pressure difference of 6-7 ~~decades~~ orders of magnitude or more across the membrane. Compared with compositions known in the prior art, these compositions offer improved resistance to kinetic decomposition and reduced chemical expansion, as well as improved stability against reduction to metal and reaction with carbon dioxide and water.

Claims:

(amended)

1.   A solid multicomponent membrane for use in a reactor wherein ~~characterised in that~~

the membrane comprises a mixed metal oxide having a structure represented by the formula:

$$La_{1-x}Ca_x(Fe_{1-y-y'}Ti_yAl_{y'})_wO_{3-d}$$
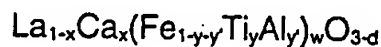
wherein x, y, y', w, and d each represent a number such that $0.1 \leq (y+y') \leq 0.8$, $0.15 \leq (x+y') \leq 0.95$, $0.05 \leq (x-y) \leq 0.3$, $0.95 < w < 1$, and d equals a number that renders the compound charge neutral and is not less than zero and not greater than about 0.8.

(Amended) The

2.   ~~A~~ membrane according to claim 1, wherein ~~characterised in that~~

the x, y, y', w, and d each represent a number such that $0.15 < (y+y') < 0.75$, $0.2 < (x+y') < 0.9$, $0.05 < (x-y) < 0.15$, $0.95 < w < 1$, and d equals a number that renders the compound charge neutral and is not less than zero and not greater than about 0.8.

(Amended) The

3.   ~~A~~ membrane according to claim 1, wherein ~~characterised in that~~

$0 < y < 0.75$ and $0 < y' < 0.3$.

amended 4.   Use of the membrane according to claims 1- 3, in a reactor for generating heat by oxidation of a carbon containing fuel to CO_2 and H_2O on the oxidation side of the membrane reactor.

debris glacier n : a glacier composed of ice that has fallen in fragments from a larger and higher glacier

debris-slide \...-ˌslīd\ n : a mass of predominantly unconsolidated and incoherent soil and rock fragments that has slid or rolled rapidly down a steep slope when comparatively dry to form an irregular hummocky deposit — compare DEBRIS-AVALANCHE

de Bro·glie equation \də-ˈbrōglē, də-ˈbrȯ-ye-\ n, usu cap B [after Prince Louis V. de Broglie] : an equation in physics: the de Broglie wavelength of a moving particle is equal to the Planck constant divided by the momentum of the particle

de Bro·glie wave n, usu cap B [after Prince Louis V. de Broglie] : the hypothetical wave train that in the wave-mechanical theory corresponds to a moving elementary particle (as an electron or proton), moves with it, and gives the particle certain wave properties (as interference and diffraction)

de·bruise \də-ˈbrüz, dē-\ vt [ME debrusen to break to pieces, fr. OF debruiser, fr. de- + bruiser to break — more at BRUISE] 1 of a heraldic ordinary : to cross or partly cover (a coat of arms or charge) as if laid over 2 of a person : to assume or use a heraldic ordinary that crosses or partly covers (a coat of arms or a charge)

debs pl of DEB

1debt \ˈdet, usu -ed-+V\ n -s [ME dette, alter. (influenced in spelling) by MF debte, fr. OF, alter. — influenced in spelling by L debitum — of dette, fr. OF dette, fr. (assumed) VL debita, fr. L, pl. of debitum debt, fr. neut. of debitus, past part. of debēre to owe, fr. de- + habēre to have — more at DE, HABIT] 1 : a neglect or violation of duty : FAULT, SIN, TRESPASS (forgive us our ~s —Mt 6:12 (RSV)) 2 : a state of owing (hopelessly in ~) 3 : something (as money, goods, or services) owed by one person to another (a mortgage ~) : something that one person is bound to pay to another or perform for his benefit : something owed : OBLIGATION (~ of gratitude) 4 : the common-law action for the recovery of a certain specified sum of money or for a sum due or of a sum that can be simply and certainly ascertained — called also action of debt; compare FORM OF ACTION

decarbonate de·car·bon·ate \(ˈ)dē + \ vt [ISV de- + calcify] : to remove calcium or calcium compounds from

de·cal·co·ma·nia \di-ˌkalkə-ˈmānē-ə\ n -s [F décalcomanie, fr. décalquer to copy by tracing, fr. dé- + calquer to trace; ...]

de·cane \ˈdek-ˌān\ n -s [ISV decane + -ol] : any of the decyl alcohols derived from normal decane

de·ca·nol \ˈdekə-ˌnȯl, -ˌnōl\ n -s [decane + -ol] : any of the decyl alcohols derived from normal decane

1decant \di-ˈkant, dē-\ vt [F décanter, fr. ML decanthare, fr. L de- + ML canthus edge, rim] 1 : to pour (as wine) from one vessel into another container 2 : to draw off (a liquid) without disturbing the sediment or precipitate

2decant n -s [decant + -ant] : the liquid decanted

de·cant·er n -s : one that decants